Technical Requirements for the Control of ^{129}I in a Nuclear Fuels Reprocessing Plant

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November 1979

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SUMMARY

A proposed regulation would limit the release of ^{129}I to the environment to 0.005 Ci/GWe-yr of nuclear power produced. This corresponds to an overall ^{129}I retention factor of about 250 for the LWR fuel cycle.

Technologies available and under development for removing iodine from off-gas and waste water streams in an LWR nuclear fuel reprocessing plant and for converting iodine to stable forms and retaining it for interim and long-term storage, transportation and isolation are discussed. Possible developments are reviewed.

Technologies available for retaining iodine from off-gas and excess water streams appear adequate, when properly combined, to meet the proposed regulation in an LWR fuel reprocessing plant, though additional work on solid sorbents is needed. Demonstrations under plant conditions are needed to define and resolve problems not met in laboratory experiments and to define practical operating conditions.

Criteria for storage and disposal forms for retained iodine are needed to guide effective development of such forms. Because no disposal form for ^{129}I is likely to survive in any environment for even one half-life (1.6×10^7 yr), work on a disposal form should seek forms that disintegrate as slowly as possible to maximize dilution before the isotope reaches the biosphere.

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TECHNICAL REQUIREMENTS FOR THE CONTROL OF ^{129}I IN A NUCLEAR FUELS REPROCESSING PLANT

INTRODUCTION

Spent nuclear fuels, when reprocessed to recover their valuable fissile content, contain fission product iodine isotopes, both stable and radioactive. These iodine isotopes are ^{127}I (stable), ^{129}I (half-life of 1.6×10^7 yr) and ^{131}I (half-life of 8 days). In spent fuels aged 150 to 200 days before reprocessing, as is customary for light water reactor (LWR) fuels, the short-lived ^{131}I has already decayed to such a low level that risk to the biosphere would be negligible even if it were all released to the environment. However, the long-lived ^{129}I is present in amounts great enough to represent significant risk to the biosphere if released.

A recently published regulation⁽¹⁾ would limit the release of ^{129}I to 0.005 Ci/GWe-yr for the commercial fuel cycle. About 1.2 Ci of ^{129}I are produced in LWR nuclear fuels per GWe-yr.⁽²⁾ To meet the proposed regulation, only 0.25 of the ^{129}I produced in the LWR fuel cycle can be released to the environment. The rest must be retained, converted to stable forms, and disposed of in a manner that presents negligible risk to the environment. The proposed ^{129}I limit (referred to hereafter as the 40 CFR 190 limit) would require an overall iodine retention factor (RF) of about 250. We have assumed that a design basis RF of about 1000 is desirable to ensure meeting the proposed limit.

Considerable technology is already available for removing iodine from gas and aqueous streams and converting it to stable forms that may be suitable for storage and disposal. This report briefly describes that technology, assesses it for use in reprocessing LWR fuels, and states the authors' views on additional needs. The report is not a literature review; two literature reviews of applicable technology have been published recently by Oak Ridge National Laboratory (ORNL).^(3,4)

BACKGROUND

Figure 1 shows schematically the points at which iodine might be released to the environment, either as gas or in solution, in a typical nuclear fuel cycle. Small amounts of iodine might be released in a nuclear reactor by damaged fuel elements, but this is expected to be insignificant in the overall control of radioiodine. Large amounts could be released in a reactor accident involving extensive fuel ruptures or fuel meltdown. Special technologies and containments have been developed to meet such emergencies. They will not be discussed in this report.

Iodine released to the environment via the off-gas or cooling water in fuel element storage basins represents a very small fraction of that in the fuel. The source would be damaged fuel elements; these are normally placed in canisters to prevent release of contamination while in storage. Control of iodine in storage basins is not discussed in this report although the technology discussed could be applied to storage basins if needed.

The main place at which iodine might be released to the environment from the fuel cycle is the fuel reprocessing plant, where the fuel elements are cut open, fuel material is completely dissolved, and numerous operations are performed to recover valuable constituents, to accumulate and concentrate wastes, and to discharge excess water. The technology discussed in this report concerns primarily the control of iodine in a fuel reprocessing plant. Some iodine may be present in the high-level liquid wastes that will be stored and subsequently converted to disposable solids. If iodine evolution proves a problem in this operation, the technology developed for its control in the reprocessing plant can be applied.

At present, reprocessing is not being done in the United States. However, if this deferral were lifted, the technology for iodine control would be necessary.

Aging spent fuel elements for 150 to 200 days before reprocessing, and thus avoiding problems from ^{131}I , is practical for LWR fuels because of their low fissile material content. Such long cooling may not be practical

FIGURE 1. Release Points for Iodine

for breeder reactor fuels because they are high in fissile content and the inventory cost of long cooling may be prohibitive. It may be necessary to reprocess them at such short cooling times that release of ^{131}I must be controlled to avoid short-term problems near the plant. Higher iodine retention factors will be required than are needed to meet the 40 CFR 190 regulation for ^{129}I . Although this report does not concern the control of iodine in a breeder fuel reprocessing plant, much of the technology discussed would be applicable. The specific problem of breeders is being addressed in work at ORNL [see Reference 5 and succeeding liquid metal fast breeder reactor (LMFBR) Fuel Recycle Program Progress Reports from ORNL].

Figure 2 shows the expected path of iodine through a typical LWR fuel reprocessing plant.⁽⁶⁾ Spent fuels from the storage basin may be partially disassembled mechanically to remove non-fuel parts. The fuel elements are

Off-gas from the shearing and fuel dissolution operations, usually called dissolver off-gas (DOG), will contain most (>90%) of the iodine that was present in the fuel. Off-gases collected from the other process vessels are

usually combined to form a second gas stream, the vessel vent off-gas. It will contain a small fraction ($<10\%$) of the iodine present in the fuel. Small amounts of iodine ($\leq 0.1\%$) are expected to be present in the high-level liquid waste to be stored and ultimately solidified. Small, but unknown, amounts of iodine will be held in spent solvent, which will probably be burned as a step in its disposal. Hence the streams of concern for iodine control are the two off-gas streams and the excess water stream. Technology has been and is being developed for removing iodine from these streams.

REMOVAL OF IODINE FROM OFF-GAS STREAMS

Technology developed for removing iodine from fuel reprocessing off-gas streams has focused on three techniques: 1) mercuric nitrate-nitric acid scrubbing (Mercurex), 2) scrubbing with hyperazeotropic (>20 M) nitric acid (Iodox), and 3) sorption of iodine on solid media. These three techniques are discussed in the following sections.

Before discussion of this technology it should be noted that, in addition to elemental iodine and inorganic iodine compounds, gaseous and aqueous iodine-containing streams in a fuel reprocessing plant will contain some iodine as organic iodides. The organic iodides are formed through reaction of iodine, nitric acid and organic materials present (solvent, carbon in fuel materials, etc.) They must be considered in any iodine removal process; they often limit the iodine retention factors that can be attained in any given process.

MERCUREX PROCESS $[\text{Hg}(\text{NO}_3)_2 - \text{HNO}_3]$ SCRUBBING

The Mercurex process involves scrubbing the off-gas with a $\text{Hg}(\text{NO}_3)_2 - \text{HNO}_3$ solution in packed or bubble cap columns. Scrubbing efficiency as a function of $\text{Hg}(\text{NO}_3)_2$ and HNO_3 concentration has been studied over wide concentration ranges. Optimum performance is obtained with $\text{Hg}(\text{NO}_3)_2$ concentrations of 0.2 to 0.4 M and HNO_3 concentrations of 8 to 10 M. The higher nitric acid concentration improves organic iodide removal.

Molecular iodine reacts in the $\text{Hg}(\text{NO}_3)_2 - \text{HNO}_3$ solution to produce HgI_2 and $\text{Hg}(\text{IO}_3)_2$. Formation of iodate is prevented by the presence of NO_2 in the off-gas stream (as is the case for the dissolver off-gas). Organic iodides react to produce HgI_2 and organic nitrates or alcohols. The capacity of the $\text{Hg}(\text{NO}_3)_2 - \text{HNO}_3$ scrub to retain iodine in solution is limited by the solubility of HgI_2 . A mole ratio of mercury to iodine of at least 4 must be maintained. Thus the capacity of a 0.4 M $\text{Hg}(\text{NO}_3)_2$ scrub to retain iodine is about 13 g I_2 /liter.

Laboratory studies of the Mercurex process with packed and bubble cap columns have produced iodine RFs in excess of 10^4 for both I_2 and CH_3I . Very little engineering-scale data on the process is available. As used at the ORNL

transuranic (TRU) facility, iodine RFs of about 100 were obtained.^(7,8) Retention factors of 10 to 75 are claimed for the system designed into the LWR fuel reprocessing plant built by Allied-General Nuclear Services at Barnwell, South Carolina.⁽⁹⁾

The Mercurex process was studied extensively at ORNL. Reference 3 provides an excellent review of Mercurex technology, with numerous references to the original literature.

Unless further development of the Mercurex process at engineering scale and under plant conditions can develop operating modes ensuring iodine retention factors of $>10^3$, this process alone does not appear adequate for treatment of the dissolver off-gas stream (expected to contain $>90\%$ of the iodine which enters the plant). It could well be used to remove the bulk of the iodine in the DOG stream and be followed by a solid sorbent (e.g., Ag mordenite) for further clean-up. It might be adequate alone for treatment of the vessel vent off-gas stream because this stream is expected to contain $<10\%$ of the iodine entering the plant.

Further engineering-scale studies under plant conditions are needed to better understand and improve the process. Holladay⁽³⁾ suggests that the iodine RF attainable with the process in practice is limited by the bleed-off of iodine from recirculated scrub solution. Perhaps operating modes could be developed to minimize this effect. The technology appears adequate for practical application for intermediate iodine RFs (~ 100).

IODOX PROCESS FOR IODINE RETENTION

Hyperazeotropic ($>20\text{ M}$) nitric acid is used to scrub iodine from a gas stream in the Iodox process. This process, an innovation of ORNL, is under development there.^(3,10,11)

As the off-gas stream passes countercurrent to the scrub solution in packed or bubble cap columns, iodine accumulates in the scrub solution as HI_3O_8 . The iodine-rich scrub is further concentrated in an evaporator; distilled HNO_3 is available for recycle; and the bottoms are further

concentrated to dehydrated HI_3O_8 . The integrated system also includes a salt extraction distillation unit for maintaining the hyperazeotropic nitric acid.

Extensive tests of an integrated Iodex system have been made at ORNL, and considerable work has been done to understand the basic chemistry of the system. Iodine retention factors $>10^4$ were consistently attained for both elemental iodine and methyl iodide. These high retention factors were confirmed in a test of the bubble cap scrubber during a transuranium element processing campaign at the ORNL TRUE facility.⁽¹¹⁾

Some of the process advantages are:

- It introduces no new chemicals into the fuel reprocessing plant system.
- It gives high retention factors for either elemental iodine or methyl iodide.
- The product formed, presently considered to be HI_3O_8 , is readily transformed to an insoluble iodine compound suitable for concreting.

Some disadvantages are:

- The integrated system involves a relatively large amount of complex equipment that will be expensive to construct and will require careful operation.
- Hyperazeotropic nitric acid can be corrosive under some conditions. For protection against corrosion, expensive construction materials will be required.
- The possibility exists for accumulation of explosive organic nitrates in scrubber bottoms and condensates.

The Iodex process alone appears capable of producing adequate iodine RFs when treating the dissolver off-gas stream, to meet the 40 CFR 190 regulation in a LWR reprocessing plant. Further engineering-scale tests under plant conditions are needed. More data on corrosion are needed to aid in selecting construction material and to define process operating limits. The potential organic nitrates need evaluation to define process operating limits.

SOLID SORBENTS FOR IODINE REMOVAL

Many solid materials can effectively remove iodine from gas streams. Solid sorbents generally offer several advantages over liquid scrubbing systems: they usually involve less complicated equipment, which is less expensive and simpler to operate; they involve less corrosive conditions; and the waste product is dry and easily handled. Some disadvantages are relatively high operating costs if the sorbent is used on a once-through basis, and solid sorbents are more sensitive to operating conditions than are liquid scrubbing systems.

Activated carbon has been studied extensively for removing iodine from gas streams, particularly in connection with nuclear power plants. It is not a candidate for iodine removal in Purex-type nuclear fuel reprocessing plants, where off-gas streams generally contain some oxides of nitrogen. Nitrogen oxides not only adversely affect (i.e., decrease) retention of iodine by activated carbon beds, but they can also lead to explosive reactions in the beds.⁽¹²⁾

Iodine also deposits from gas streams onto many other solids such as sand, soil, and wall coatings and in tubes and ducts. These phenomena are of interest from the standpoint of contamination and contribution to errors in measurement, but they are not of use for high-efficiency removal of iodine from off-gas streams.

Solid sorbents currently receiving most attention for removing iodine from gas streams are metal-exchanged zeolites and silver-doped gels and oxides (alumina and silica). These materials have the advantages of being nonflammable, of operating well at elevated temperatures, of having relatively high capacity for iodine sorption, of producing good (10^2 - 10^3) iodine and organic iodide RFs under proper operating conditions, and of producing a dry solid iodine product which may serve directly as an iodine storage and disposal form. Solid sorbents, with and without additives, have been investigated extensively by workers at the Idaho National Engineering Laboratory (INEL). Excellent literature reviews of the technology of solid iodine sorbents are given in References 3 and 4.

Silver-Doped Gels and Oxides

Silver and other metals and their salts have been studied as coatings on many inert solids. Work at INEL and ORNL showed that a variety of combinations were effective in removing iodine from gas streams. The most effective additive was silver, and most of the subsequent studies have used silver compounds on porous alumina or alumina-silica.

Silver-containing alumina and alumina-silica combinations were evaluated for removing iodine from gas streams by workers at ORNL.⁽¹³⁾ These materials have trade designations:

Gx-100	alumina	8 x 16 or 16 x 30 mesh
Gx-135	alumina-silica	8 x 16 mesh
Gx-148	alumina-silica	16 x 30 mesh

The Environmental Protection Agency (EPA) has sponsored work at the University of Belgrade, Yugoslavia, on the capture of methyl iodide using various metal nitrates on Alcoa alumina H-151.⁽¹⁴⁾ The base sorbent composition is 90% Al_2O_3 , 1.4% Na_2O , 0.1 Fe_2O_3 , and 1.7% SiO_2 . Tests have been made with the base sorbent impregnated with the following metal nitrate: Ag, Cu, Pb, Cd, Zn, Ni, Co, Ce, Ag + Cu, Ag + Ce, Ag + Cd. Only the silver-doped sorbent and, to a lesser extent, copper-, lead- and cadmium-doped sorbent showed irreversible chemisorption of CH_3I at temperatures below 100°C.

These silver-loaded solids appear capable of high iodine retention factors ($>10^3$) for both elemental iodine and organic iodides. However, they appear to have deficiencies under process conditions, e.g., in the presence of contaminant gases, and their development has not been pursued.

Much more extensive has been the work with a silver-nitrate-impregnated amorphous silicic acid, commonly called AC-6120. It has been developed in the Federal Republic of Germany as a nonregenerable solid sorbent for iodine.⁽¹⁵⁾ Silver content is from 7 to 12 g/100 g sorbent. Its performance in the presence of other gases has been demonstrated. It has been used successfully to remove iodine from dissolver off-gas at the Karlsruhe Reprocessing Pilot Plant and at the SAP Marcoule plant.⁽¹⁶⁾

Metal-Exchanged Zeolite Sorbents

Zeolite materials have a wide range of properties, depending on their composition.⁽¹⁷⁾ An important physical characteristic is the pore size. An important chemical characteristic is the $\text{SiO}_2/\text{Al}_2\text{O}_3$ mole ratio. Early iodine studies were performed with Types X and Y zeolites, properly classified as faujasites. They have a low (<10) $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio. The faujasites have a high exchange capacity but a low resistance to acids and a low temperature stability. More recently interest has increased in the mordenites. They have a high (>10) $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio and are more stable thermally and towards acids. They do, however, have a lower exchange capacity than the faujasites.

Extensive studies have been made of retention of iodine from gas streams by metal-substituted zeolites.^(3,4,18,19) These have concerned the different types of zeolites, physical form of the zeolite, substituted metals and their oxidation states, loading capacity, temperature effects, effects of other gases and methods for regenerating the zeolites for reuse.

Of all the metal-substituted zeolites studies, only silver-substituted sorbers show promise for consistently high RFs for I_2 and CH_3I under practical operating conditions for a fuel reprocessing plant. Retention factors >100 are readily obtained with the silver-substituted zeolites at iodine loadings that are practical for plant application.

The chief disadvantage of silver zeolites is the cost of the silver if the beds are used on a once-through basis. A possible way to avoid part of this cost would be to use a cheaper but less efficient zeolite bed ahead of the silver-substituted bed. Most of the iodine would be retained on the cheaper zeolite; the expensive silver zeolite would provide a high overall retention factor but load to exhaustion much slower. Copper, zinc, and cadmium zeolites were prepared, but brief studies indicated that iodine sorption was physical and not chemical and that the iodine loadings obtained were poor.^(18,19)

Another approach to reducing the operating cost for silver-substituted iodine sorbents is to regenerate them (remove the iodine) and reuse them. The removed iodine could then be resorbed onto a cheaper sorbent (e.g., lead zeolites) for storage or disposal, or the iodine could be captured in aqueous

media, converted to an insoluble iodine compound, and concreted. Studies at INEL showed that the sorbents could be regenerated using dry H_2 at 400 to 500°C. Iodine is removed as hydrogen iodide, and the silver in the sorbent is reduced to metallic silver. However, silver zeolites tend to lose exchange capacity with succeeding regenerations.⁽¹⁹⁾ In this respect, the mordenite structure is superior to the faujasite structure. Regeneration tests on silver faujasite showed a twofold reduction in capacity after 5 cycles,⁽¹⁵⁾ while silver mordenite has been regenerated 13 times with about 20% loss of capacity.⁽¹⁹⁾

Although the silver-impregnated alumina and silica sorbers and the silver-substituted zeolites appear capable of producing iodine retention factors in excess of 10^3 , some are sensitive to process conditions (humidity, NO_2). Again, the mordenite is superior to the faujasite because of better acid resistance. It is uncertain whether at this stage of technical development they could be depended on alone to give, consistently, the 10^3 retention factor needed for the dissolver off-gas stream to meet the 40 CFR 190 requirement. They could be used effectively for iodine clean-up following Mercurex scrubbing or alone on the process vessel off-gas stream.

Regeneration and reuse of the silver-substituted mordenites appears promising as a means of reducing operating cost by conserving silver. However, it introduces process design complications and raises safety issues.

Further laboratory work toward better understanding of the nature of metal-zeolite-iodine reactions and the effects of organics and organic iodides on sorbent performance is needed. More information on the effects of contaminant gases is also needed. studies in this area are continuing at Pacific Northwest Laboratory (PNL).⁽²⁰⁻²²⁾ Engineering-scale demonstrations under plant conditions are needed to optimize operating conditions and lend confidence in sorbent performance.

SUMMARY OF IODINE RETENTION TECHNOLOGY

Of the processes already developed for removing iodine from off-gas streams, only the Iodox process appears capable of consistently

producing the iodine RFs of $\sim 10^3$ needed to meet the 40 CFR 190 regulation for control of ^{129}I . The Iodox process involves costly and complicated equipment and will require close process control. It has inherent corrosion problems and may involve problems with accumulation of explosive organic nitrates. It produces a product that must be converted to a more stable form for long-term storage or disposal.

The Mercurex process involves less costly and complicated equipment than the Iodox process. Corrosion problems should be less severe than with Iodox, and it should be easier to maintain proper operating conditions. It also produces a waste product that must be converted into a more stable form for storage and disposal. In addition, mercury, a chemical not used elsewhere in a Purex-type fuel reprocessing plant, is introduced into the process. Mercury is a toxic substance that will volatilize under some plant conditions. Furthermore, it is expensive as well as being a valuable resource for other uses. While procedures have been explored for recovering and reusing the mercury, these have not yet proven operable and would require additional equipment and operations in the plant.

Solid sorption processes are the simplest of the three processes in terms of equipment and operation. No new chemicals are introduced that could get dispersed throughout the plant. No further processing of the waste product is required if, as is often assumed, the solid sorbents represent an acceptable waste storage or disposal form. No problems of corrosion or chemical instability are inherent in the process. Iodine retention factors attained are near those needed for the 40 CFR 190 limit even when applied to the dissolver off-gas system.

For these reasons continued development of the solid sorbent systems to define conditions that will ensure consistently adequate iodine retention is highly desirable. Also, further work should be done to define combinations which will give adequate iodine retention, incur reasonable cost, and avoid, if possible, the regeneration step. Hydrogen reduction for the regeneration step is, at best, hazardous. Some effort to find other iodine removal techniques is desirable.

REMOVAL OF IODINE FROM EXCESS WATER

Excess water is simply that water which cannot be recycled within the fuel reprocessing plant. It is generally the condensate from waste evaporation and will usually contain a small amount of HNO_3 ($\sim 0.01 \text{ M}$). Part of the iodine present may be organic iodides. Excess water and the off-gas streams are the only effluents that will be discharged routinely to the environment from a fuel reprocessing plant.

Whether the excess water will have to be treated for iodine removal to meet the 40 CFR 190 regulation is not known at this time. The amount of iodine expected in excess water has been estimated for the AGNS plant at Barnwell, South Carolina,⁽⁹⁾ and for the proposed EXXON Nuclear Company plant.⁽²³⁾ These estimates meet, or nearly meet, 40 CFR 190 requirements. However, if they err on the low side, treatment of the excess water for iodine removal will be required, and it is desirable that the technology be available. At any rate, it is unlikely that iodine retention factors greater than 10 to 100 will be needed.

Two approaches for removing iodine from excess water have been studied: sorption of the iodine in excess water onto solid sorbents, and prevention of iodine evolution during waste concentration.

RETENTION OF IODINE BY SOLID SORBENTS

Early work on the sorption of iodine from excess water by solid sorbents was done at ORNL using Amberlite® XAD-12.⁽²⁴⁾ Later work was done at PNL⁽⁶⁾ using other Amberlite XAD macroreticular polymeric sorbents (XAD-12 is no longer commercially available) and also anion exchange resins.

The macroreticular polymeric sorbents do not function as ion exchangers but sorb iodine as molecular iodine. Early PNL work with these sorbents showed poor iodine retention from dilute nitric acid containing elemental iodine,

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presumably due to reduction of iodine to iodide by the sorbent. Further studies then involved addition of H_2O_2 to the excess water ($0.01 \text{ M HNO}_3 - 10^{-5} \text{ M I}_2$) to counteract the reducing properties of the sorbent. Results were promising (iodine retention factors in the range >10 to >100 for >1000 column volumes of feed). Of the sorbents studied, XAD-4 appeared the best.

As an alternative to adding H_2O_2 to the excess water, further work at PNL studied the use of sorbents pretreated with an oxidant. Pretreatment of the polymeric sorbents with KMnO_4 solutions produced sorbents that showed good retention factors for iodine from excess water. Whether the iodine was present initially as molecular iodine or as iodide ion had little effect on sorbent performance. Iodine retention factors from 10 to >100 for feed throughputs of >3000 column volumes were obtained. MnO_2 deposited on these sorbents gradually washed off during the iodine loading operation. Washing the iodine off the sorber with NaOH and NaOH -ethanol solutions and regenerating the columns with KMnO_4 treatment has been studied, but not adequately to define optimum procedures.

Retention of iodine from excess water by Amberlite anion exchangers, particularly Amberlite IRA-900, was studied at PNL. While this sorbent normally operates by ion exchange, both elemental iodine and iodide ion were efficiently retained. Whether the molecular iodine was retained by surface sorption or by the sorber's reducing iodine to iodide and retaining it as iodide is not entirely clear. Iodine retention factors of 10 to >100 for feed throughputs up to ~ 500 column volumes were obtained. Essentially complete elution of retained iodine by about 30 column volumes of $1 \text{ M HNO}_3 - 0.1 \text{ M hydroxylamine nitrate}$ was attained when the iodine was sorbed as iodide. Elution by single-solution was not effective when the iodine was sorbed as molecular iodine. Complete elution was accomplished using alternate washes of 1 M NaOH and 1 M HNO_3 totaling about 100 column volumes. The exchanger could be recycled repeatedly without loss of iodine retention factor or capacity.

Another alternative would use a silver zeolite in the evaporator off-gas. There is little doubt but that this would be adequate. The disadvantage is that the gas flow would be very large, in excess of 10^5 m^3 per day. A large bed and additional heating would be required.

PREVENTING IODINE VOLATILIZATION DURING WATER EVAPORATION

Two approaches to suppressing volatilization of iodine during evaporation processes have been studied.⁽⁶⁾ In the first an iodine complexing agent such as mercuric nitrate is added to the solution being evaporated. Iodine retention factors (bottoms) of up to 5000 have been obtained in laboratory equipment simulating volatilization of excess water ($0.01 \text{ M HNO}_3 - 10^{-6} \text{ M I}_2$). Bottoms composition was $0.5 \text{ M HNO}_3 - 0.01 \text{ M Hg (NO}_3)_2 - 10^{-4} \text{ M I}_2$. In this equipment, the deentrainment DF (for nonvolatile species) was about 9000.

In similar experiments in which the bottoms composition was $0.5 \text{ M NaNO}_3 - 0.05 \text{ M NaOH} - 10^{-4} \text{ M I}$ and the feed was $0.01 \text{ M HNO}_3 - 10^{-6} \text{ M I}_2$, iodine retention factors of 5000 to 8000 were obtained.

In both cases, higher retention factors were obtained when the feed was added beneath the surface. In the alkaline vaporization case, lower retention factors were obtained when the feed was made alkaline before being added to the concentrator than when acidic feed was added.

It should be noted that the effective iodine retention factor attained in vaporization of excess water (iodine concentration in feed/iodine in concentrator condensate) depends on the degree of concentration attained in the evaporator. For example, at a concentration factor of 50, a bottoms retention factor of 1500 corresponds to a feed retention factor of only 30. For a concentration factor of 100, the feed retention factor would be only 14.

SUMMARY OF REMOVING IODINE FROM EXCESS WATER

Both of the solid systems studied for removing iodine from excess water—pretreated macroreticular polymeric sorbents and anion exchange resins—appear capable of producing iodine retention factors in the range of 10 to >100 under laboratory conditions and carefully controlled operating conditions. Treatment capacity is significantly larger for the polymeric materials (~3000 column volumes) than for the anion exchange resins (~500 column volumes). Either can be regenerated for reuse if this is necessary or desirable to make the processes practical. In the case of permanganate-treated polymeric sorbents, manganese washes off the sorbent during the loading cycle and is, then, present in the water when it is discharged.

Both methods of retaining iodine in the bottoms of concentrators when evaporating excess water—addition of $\text{Hg}(\text{NO}_3)_2$ and alkaline distillation—appear capable of iodine retention factors (feed) in the 100 to 200 range for concentration factors of 50.

If present predictions of the amount of iodine that will be in excess water are accurate or nearly so, iodine retention factors provided by these systems should be ample to meet the 40 CFR 190 proposed limit for ^{129}I release. More actual plant experience backed up by more precise data on the path of iodine through a fuel reprocessing plant is needed.

All these studies are deficient in that they have been done with laboratory feedstocks instead of plant solutions and that they have been done in laboratory equipment at laboratory scale rather than in plant equipment, under plant conditions, and at plant scale. Retention factors tend to be less under plant operating conditions than under laboratory conditions. Engineering-scale demonstrations with plant-derived feedstocks are needed before applicability of the processes can be fully defined. This is particularly so for the solid sorbent systems because iodine retention appears quite dependent on the chemical nature of iodine in the feed solutions.

What to do with the waste products from any of these processes needs further consideration. If solid sorbents are used on a once-through basis, are they acceptable as is for long-term storage or disposal? If not, what further treatment is needed? If they are to be eluted and regenerated for reuse, what disposition can be made of the eluates and regeneration solutions? Can they be put into high-level wastes without adversely affecting planned treatment and disposal of these wastes? The same questions need answers for the bottoms from $\text{Hg}(\text{NO}_3)_2$ or alkaline distillation of excess water. If it is necessary to remove iodine from these solutions and to convert it to a stable form for storage or disposal, what processes are available and what new technology must be developed? Fortunately, the fraction of total iodine represented by these streams is very small and only a modest recovery may be required.

CONVERSION OF IODINE TO STABLE FORMS

A major problem in developing the technology for producing stable iodine forms for interim storage, transportation and disposal is the lack of established criteria these forms must meet. This is particularly true for disposal. The extremely long half-life of ^{129}I (1.6×10^7 yr) makes it unlikely that any chemical form of iodine will persist in any environment for even one half-life of ^{129}I . Total confinement until ^{129}I has decayed to insignificance appears impossible to ensure. Perhaps forms that will meet the needs for interim storage and transportation to a disposal site are more significant at present than a form specifically designed for disposal. At any rate, criteria for the stable form are needed to guide research and development toward producing such forms and providing technology for converting retained iodine to the forms.

SOLID SORBENT FORMS

Solid sorbents for iodine such as may be used in off-gas or excess water streams are often considered an acceptable form for storage or disposal without further treatment. Such a decision cannot be made until criteria are developed and accepted for iodine disposal forms. Little or no data have been produced on the stability of these iodine-loaded sorbents in regard to leaching by water (or brine), long-term heating, and other properties usually defined for candidate disposal forms. If they are to be seriously considered as disposal forms, these data must be obtained.

If silver-substituted zeolites are regenerated for reuse, two routes may be followed. In one the eluted iodine may be resorbed on a less expensive sorbent such as lead-substituted zeolite. If the lead zeolites are to be considered a disposal form, stability data are needed for them. In the other route, the eluted iodine, as hydrogen iodide, could be scrubbed into an aqueous system and subsequently converted to a disposal form (e.g., converted to an insoluble iodine compound and incorporated into a stable matrix such as cement). Some data are available on this approach and work is continuing on it. (21)

CEMENTED FORMS

Some work has been done on converting the iodine retained in aqueous solutions to stable forms. Major emphasis has been toward converting iodine to insoluble compounds and incorporating these compounds in cement. Iodic acid, the product obtained in the Iodex process, can be dissolved in water and precipitated as an insoluble iodate— $\text{Ba}(\text{IO}_3)_2$ or $\text{Hg}(\text{IO}_3)_2$ —which can then be cast in cement.

The Mercurex waste product presents a more difficult problem. Direct concentration of the $\text{Hg}(\text{NO}_3)_2 - \text{HgI}_2 - \text{HNO}_3$ solution results in volatilization of some iodine and conversion of the remainder to slightly soluble $\text{Hg}(\text{IO}_3)_2$. Concentration of the solution by introducing it into concentrated (15.3 M) HNO_3 bottoms avoids significant iodine evolution. Iodine retention factors (feed/condensate) of 10^3 to $>2 \times 10^4$ were obtained.⁽²⁵⁾

Concreting the entire $\text{Hg}(\text{IO}_3)_2 - \text{Hg}(\text{NO}_3)_2 - \text{HNO}_3 - \text{H}_2\text{O}$ slurry can be done. Removal of the $\text{Hg}(\text{IO}_3)_2$ solid for concreting and return of the residual $\text{Hg}(\text{NO}_3)_2 - \text{HNO}_3$ solution to process is also possible. This not only conserves mercury but also reduces the waste volume. This technology, however, has not been demonstrated. A problem encountered here is the slow precipitation of $\text{Hg}(\text{IO}_3)_2$ from the solution. Dilution of the filtrate to prevent deposition of $\text{Hg}(\text{IO}_3)_2$ in process equipment has been suggested.⁽²⁵⁾

Mercury is a relatively expensive reagent and a valuable resource for other uses. It is also a highly toxic material that must be disposed of under controlled conditions. Hence it has been suggested that the $\text{Hg}(\text{IO}_3)_2$ be treated with NaOH to produce solid HgO and NaIO_3 in solution. The HgO can be separated, dissolved in HNO_3 , and returned to the scrubber solution. The iodine can be converted to another insoluble iodate such as $\text{Sr}(\text{IO}_3)_2$ or $\text{Ba}(\text{IO}_3)_2$ and incorporated in cement. These processes have to date received only token study; thus considerable laboratory and demonstration work needs to be done before they can be considered ready for plant application.

Incorporating iodine compounds in cement produces a solid relatively stable to mechanical shock, to leaching by water and to temperatures not exceeding the dehydration temperature of cement. The cement can be cast directly

into 55-gal drums for storage and disposal. Decay heat generated by ^{129}I in these solids would be negligible; the solids would remain essentially at ambient temperature in the disposal site.

Some consideration has been given to choice of iodine compounds (metal iodides and iodates) based on thermal stability, stability towards oxidation, solubility in aqueous systems, and cost and availability of the metal. Both theoretical and experimental studies have been carried out.⁽²⁶⁾ In the absence of criteria for storage or disposal, a choice cannot be made, but it appears that a number of compounds are candidates including compounds of copper, strontium, barium, silver, lead, cerium, mercury, and thorium. Silver is outstanding except for cost. Toxicity may eliminate mercury and perhaps lead, depending on the disposal site chosen.

Some work on the leaching of iodine from cement by water has been done at ORNL,⁽²⁷⁾ and work is currently in progress at PNL to define the influence of a series of factors on the leach rate.^(20,22,28) These factors include temperature, cement additives, iodine form, e.g., metal iodides and metal iodates, solubility of iodine compound, the concentration and surface treatment of the cement, and leachate composition and change rate. The preliminary results indicate that modifications to the cement may help to reduce the iodine release rate. However, even the best cement composite will probably permit leaching of one-half of the iodine in a few hundred years. Cement coatings and tertiary barriers can extend the time only a negligible amount compared to the half-life of ^{129}I . The question really becomes one of how fast the environment can assimilate the iodine through dilution. Again, this is related to the criteria problem. Disposal criteria based on dispersion and dilution have been studied at PNL.⁽²⁹⁾

Leach rates and stability studies of other potential solid disposal forms including glasses, zeolites, and organic compounds and composite materials should be carried out. These studies should be coordinated with the Waste/Rock Interaction (WRIT) program.

SUMMARY AND CONCLUSIONS

Techniques currently being developed for removing iodine from off-gas and excess water streams appear adequate to permit a nuclear fuels reprocessing plant to meet the proposed limit for release of ^{129}I to the environment. Properly combined, the techniques could probably permit meeting a limit one order of magnitude lower than the proposed limit. In general, the techniques lack demonstration and development under actual plant conditions.

Iodox appears adequate, alone, for treatment of dissolver off-gas, but it will involve complicated equipment and operating conditions. Continued study of the Mercurex process is needed as a back-up even if it must be used in combination with solid sorbents for the DOG stream.

Solid sorbent systems for off-gas streams are certainly simpler to operate than either Iodox or Mercurex. Because they come so near providing the iodine retention needed to meet the proposed ^{129}I release limit, work should be continued toward improving their performance to the point where they could be used alone for DOG. This should include basic studies on reaction mechanisms as well as engineering-scale demonstration under plant conditions.

Further work may be needed on sorption of iodine from excess water by solid sorbents. Demonstrations with plant-derived feedstocks are needed to define iodine forms present and their sorption behavior.

The timing of the need for this technology depends on the government's future attitude toward construction and operation of nuclear fuel reprocessing plants. If the present deferral of fuel reprocessing continues, the technology will not be needed in this country. If the deferral were lifted, the technology would be needed immediately for use in the design of future fuel reprocessing plants.

Lack of accepted criteria for an iodine disposal form is a serious deterrent to effective development of disposal forms and the technology for producing such forms. There are no specific goals to aim for. A concerted effort to develop acceptable criteria for iodine (^{129}I) disposal is needed. Account

should be taken of the futility of trying to devise a disposal form that will be stable until ^{129}I has decayed to an insignificant level.

Without such criteria and without firm decisions on where and how other radioactive wastes will be disposed of, it can only be assumed that ^{129}I will be disposed of in a remote geologic formation. The threat of dispersal is then primarily one of water entering the formation. A disposal form with good resistance to leaching of iodine by water will help delay dispersal of iodine, aid in ensuring that it is highly diluted, and minimize the chance that it will appear anywhere in the biosphere in hazardous concentrations. In general, dispersal will be delayed also by the geology of the disposal site.

Work should continue on conversion of retained iodine to water-insoluble solids, on incorporating these into stable impervious matrices such as cement, on understanding the processes by which iodine is leached from such solids, and on reducing the rate at which iodine is leached. Future establishment of criteria for iodine disposal may make a higher order of leach resistance unnecessary. However, it would be premature at present to assume that the observed leach rates are tolerable. Coordination between these studies and the WRIT studies should be maintained.

The timing for developing an acceptable disposal form for ^{129}I is less critical than the timing for retention technology. Long-term interim storage of iodine recovered in a fuel reprocessing plant should not be difficult if the iodine is free of active heat-producing isotopes. Consideration should be given to safe and convenient forms for interim storage and to interim storage forms which will not subsequently involve unnecessarily difficult conversions to a disposal form.

Iodine present in spent solvent could present a release problem if and when such solvent is burned as a step in its disposal. Iodine volatilized may not be in a chemical form that is readily retained by available techniques because of the organic material present. Available data suggest that both Iodex and silver zeolites will probably achieve this removal, but it has not been demonstrated. New technology may be required for iodine retention in this operation.

Present predictions are that a negligible amount of iodine will be present in high-level waste. If this is incorrect and significant amounts of iodine must be dealt with during high-level waste treatment, available iodine retention technology should be applicable.

REFERENCES

1. Code of Federal Regulations, Title 40: Protection and Environment; Chapter 1, Subchapter F, Part 190: Environmental Radiation Standards for Nuclear Power Operations (40 CFR 190) (Revised July 1, 1977).
2. Removal, Storage and Disposal of Gaseous Radionuclides from Airborne Effluents, IAEA-209, Report of a Technical Committee Meeting held in Vienna, Austria, September 20-24, 1976.
3. D. W. Holladay, A Literature Survey: Methods for the Removal of Iodine Species from Off-Gases and Liquid Waste Streams of Nuclear Power and Fuel Reprocessing Plants With Emphasis on Solid Sorbents, ORNL/TM-6350, Oak Ridge National Laboratory, Oak Ridge, TN, January 1979.
4. R. T. Jubin, A Literature Survey of Methods to Remove Iodine from Off-Gas Streams Using Solid Sorbents, ORNL/TM-6607, Oak Ridge National Laboratory, Oak Ridge, TN, March 1979.
5. O. O. Yarbrow, D. J. Crouse, and G. I. Cathers, "Iodine Behavior and Control in Processing Plants for Fast Reactor Fuels," Proceedings of the Eleventh AEC Air Cleaning Conference, Richland, WA, August 31-September 3, 1979, USAEC Report CONF-700816 (Vol. 2), pp. 551-566.
6. J. L. Swanson, Control of Radioiodine in the Excess Water Stream of a Nuclear Fuels Reprocessing Plant, PNL-2080-9, Pacific Northwest Laboratory, Richland, WA, May 1978.
7. O. O. Yarbrow, G. I. Cathers et al., LMFBR Fuel Cycle Studies Progress Report for September 1969, No. 7, ORNL/TM-1748, Oak Ridge National Laboratory, Oak Ridge, TN, 1969.
8. O. O. Yarbrow, G. I. Cathers et al., LMFBR Fuel Cycle Studies Progress Report for February 1979, No. 12, ORNL/TM-2918, Oak Ridge National Laboratory, Oak Ridge, TN, 1979.
9. Final Safety Analysis Report - Barnwell Nuclear Fuel Separations Facility, Amendment 7, Docket No. 50-332, Allied-General Nuclear Services, Barnwell, SC, November 1975.
10. R. J. Ginnow and W. E. Clark, Iodine Recycle Studies Scoping Studies of the Solubility of Iodic Acid and of Vapor-liquid Equilibria in the System; 60 to 80 $\text{Mg}(\text{NO}_3)_2$ - 0.20 HNO_3 - 20 to 40 H_2O , ORNL/TM-6398, Oak Ridge National Laboratory, Oak Ridge, TN, December 1978.
11. E. D. Collins and D. E. Benker, Iodine Process Tests in a Transuranium Element Production Campaign, ORNL/TM-6182, Oak Ridge National Laboratory, Oak Ridge, TN, June 1979.

12. W. A. Rogers and S. L. Reese, "The Removal of Iodine from Reprocessing Plant Effluents," Reactor Fuel Processing Technology, 12:173, 1969.
13. R. D. Ackley and J. Combs, Applicability of Inorganic Sorbents for Trapping Radioiodine from LMFBR Fuel Reprocessing Off-Gas, ORNL/TM-4227, Oak Ridge National Laboratory, Oak Ridge, TN, 1973.
14. I. J. Gal et al., "Adsorption of Methyl Iodide on Impregnated Alumina," Proceedings of the 13th AEC Air Cleaning Conference, CONF-740807, p. 815, 1974.
15. J. G. Wilhelm and H. Schuttelkopf, "An Inorganic Adsorber Material for Off-Gas Cleaning in Fuel Reprocessing Plants," Proceedings of the 12th AEC Air Cleaning Conference, CONF-720823, p. 540, 1972.
16. J. G. Wilhelm, J. Furrer and E. Schultes, "Head-end Iodine Removal from a Reprocessing Plant with a Solid Sorbent," Proceedings of the 14th ERDA Air Cleaning Conference, CONF-760822, p. 447, 1977.
17. D. W. Breck, Zeolite Molecular Sieves, John Wiley and Sons, New York, NY, 1974.
18. D. T. Pence, F. A. Duce, and W. M. Maerk, "Developments in the Removal of Airborne Iodine Species with Metal Substituted Zeolites," Proceedings of the 12th AEC Air Cleaning Conference, CONF-720823, p. 417, August 1972.
19. B. A. Staples, L. P. Murphy, and T. R. Thomas, "Airborne Elemental Iodine Loading Capacities of Metal Zeolites and a Dry Method for Recycling Silver zeolite," Proceedings of the 14th ERDA Air Cleaning Conference, CONF-760822, p. 636, August 1976.
20. L. L. Burger, Nuclear Waste Management Quarterly Progress Report July Through September 1978, A. M. Platt and J. A. Powell, comps., PNL-2378-3, Pacific Northwest Laboratory, Richland, WA, January 1979.
21. L. L. Burger, Nuclear Waste Management Quarterly Progress Report October Through December 1978, A. M. Platt and J. A. Powell, comps., PNL-2378-4, Pacific Northwest Laboratory, Richland, WA, March 1979.
22. L. L. Burger, Nuclear Waste Management Quarterly Progress Report January Through March 1979, A. M. Platt and J. A. Powell, comps., PNL-3000-1, Pacific Northwest Laboratory, Richland, WA, May 1979.
23. Preliminary Safety Analysis Report - Nuclear Fuel Recovery and Recycling Center, Revision 1, Docket 50-564, Exxon Nuclear Company, Inc., September 1977.
24. Chemical Technology Division Annual Progress Report for Period Ending March 31, 1971, ORNL-4682, Oak Ridge National Laboratory, Oak Ridge, TN, 1971.

25. J. A. Partridge and G. P. Bosuego, Waste Management of Mercurex Nitrate Off-Gas Scrubber Solution, A Laboratory Study, HEDL-TME 79-11, Hanford Engineering Development Laboratory, Richland, WA, July 1979
26. L. L. Burger, Nuclear Waste Management Quarterly Progress Report October Through December 1977, A. M. Platt and J. A. Powell, comps., PNL-2377-4, Pacific Northwest Laboratory, Richland, WA, January 1978.
27. W. E. Clark, C. T. Thomas, and W. B. Howerton, Fixation of Radiodine with Portland Cement, Part I: Preliminary Scoping Studies, ORNL-TM-5064, Oak Ridge National Laboratory, Oak Ridge, TN, 1975.
28. L. L. Burger, Nuclear Waste Management Quarterly Progress Report April Through June 1978, A. M. Platt and J. A. Powell, comps., PNL-2378-2, Pacific Northwest Laboratory, Richland, WA, November 1978.
29. L. L. Burger, Nuclear Waste Management Quarterly Progress Report April Through June 1979, A. M. Platt and J. A. Powell, comps., PNL-3000-2, Pacific Northwest Laboratory, Richland, WA, September 1979.

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